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14. ABSTRACT

Presolar dust grains predate the formation of the solar system, originating from circumstellar outflows and supernova ejecta. The two most abundant forms of presolar grains isolated from primitive meteorites are nm-sized diamond1 and mm to submm- sized SIC2. Both are ubiquitous in primitive chondritic meteorites at 300 - 1800 ppm (diamond) and 1 - 28 ppm (SiC)3. Silicon carbide is particularly interesting because it is known to form hundreds of different polytype structures and the formation of a particular polytype is sensitive to growth conditions. The first astronomical evidence of SiC in dusty envelopes of carbon stars came from a relatively broad 11.3 mm infrared (IR) feature4. Later attempts to identify crystallographic structure of circumstellar SiC from IR spectra of carbon stars5-8 have generated controversy over the techniques and interpretation of the 9-13. The outstanding question of polytype has bearing on physical conditions, such as temperatures and pressures, at which SiC condense from circumstellar outflows or supernova ejecta. Therefore, using transmission electron microscopy (TEM), we unambiguously determine the distribution of polytypes in presolar SiC grains, isolated by acid dissolution from the Murchison CM2 carbonaceous meteorite.

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Polytype Distribution in Presolar SiC: Microstructural Characterization by Transmission Electron Microscopy

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Introduction:

Presolar dust grains predate the formation of the solar system, originating from circumstellar outflows and supernova ejecta. Their constituent elements are characteristic of the different nucleosynthetic processes that occurred in the different star types at various stages of stellar evolution. The two most abundant forms of presolar grains, isolated from meteorites, are nanometer-sized diamond [1] and micron- to submicron-sized SiC [2]. Both appear ubiquitous in primitive chondritic meteorites at 300 - 1800 ppm (diamond) and 1 - 28 ppm (SiC) [3].

The first astronomical evidence of SiC in dusty envelopes of carbon stars came from a relatively broad 11.3 μm infrared (IR) feature attributed to emission by small SiC particles between the transverse and longitudinal optical phonon frequencies [4, 5]. Later attempts to identify the crystallographic structure of circumstellar SiC from IR spectra [6-8] generated controversy over the techniques and interpretation of the data [9-13]. The outstanding question of polytype variation in presolar SiC has bearing on grain formation conditions, because microstructures (particularly SiC polytypes) are highly dependent on conditions and atomic-scale mechanisms of formation. Hence, microstructures archive valuable information on grain condensation mechanisms and conditions within circumstellar grain forming regions.

Discussion:

Unfortunately, there are few microstructural studies of presolar SiC. Analysis of individual 1.5 - 26 μm SiC grains from the Murchison L-series separate by Raman spectroscopy and ion probe mass spectroscopy have shown all grains exhibiting anomalous isotopic compositions were of the cubic β-SiC structure [14]. However, grains of this size are atypical, comprising less than 0.2% of the total population in number [15]. Therefore, we studied presolar SiC in the fine-grain size fraction, KJB, of the Murchison separate by transmission electron microscopy (TEM). Of the nine Murchison K-series size separates, KJB is reported to contain, the highest SiC abundance (1.91 ppm of the bulk meteorite corresponding to over 1/3 the mass of SiC in Murchison) and highest purity (97% SiC) [15]. Furthermore, KJB is a representative sampling of the total SiC population since 70% of the total population lies within 0.3 - 0.7 μm, characteristic of 90% of the grains in KJB (Figure 1). Importantly, secondary ion mass spectrometry (SIMS) measurements of individual SiC grains in KJH, KJG, KJF [16], KJE [17, 18], and KJC [19] separates indicate that nearly all (99%) are presolar mainstream grains. In all of these studies, no significant amounts of isotopically normal SiC were reported, indicating these separates contain few SiC grains that are solar nebula products or terrestrial contamination.

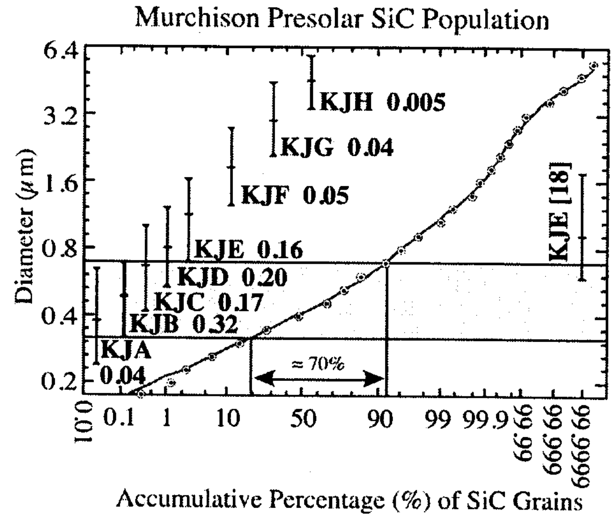

Figure 1. Murchison SiC size distribution measured by scanning electron microscopy (SEM). Dominant size range (omitting 5% tails in either end of distribution) is shown by vertical bars. Mean size is shown by the horizontal bar. The relative mass within each size fraction is also indicated.

Approximately 25% of the KJE SiC grains analyzed by SIMS [18] lie within the size range reported for KJB (Figure 1). Furthermore, there is large overlap (≈ 40 % of the total population) in grain size between the KJC and KJB separates (see Figure 1). The significant overlap for both the predominately presolar KJE and KJC with KJB strongly suggest that KJB SiC grains are also predominately presolar.

High-resolution lattice images and selected area electron diffraction (SAED) demonstrate only two SiC polytypes are present in KJB; cubic 3C (β-SiC) and hexagonal 2H (α-SiC) (Figure 2). Intergrowths of these two polytypes are frequently observed. Less common than other grain types, heavily, stacking-fault, disordered grains are also observed. Terrestrial SiC contamination can be ruled out because (i) 2H SiC has

never been reported as occurring naturally and is not found in most commercially synthesized SiC and (ii) 3C SiC is terrestrially rare in nature and also not found in most commercially synthesized SiC [20]. A nebular origin for 2H SiC in KJB can also be excluded, based on isotopic studies [16-19], if the 2H population is sufficiently large (> 1%).

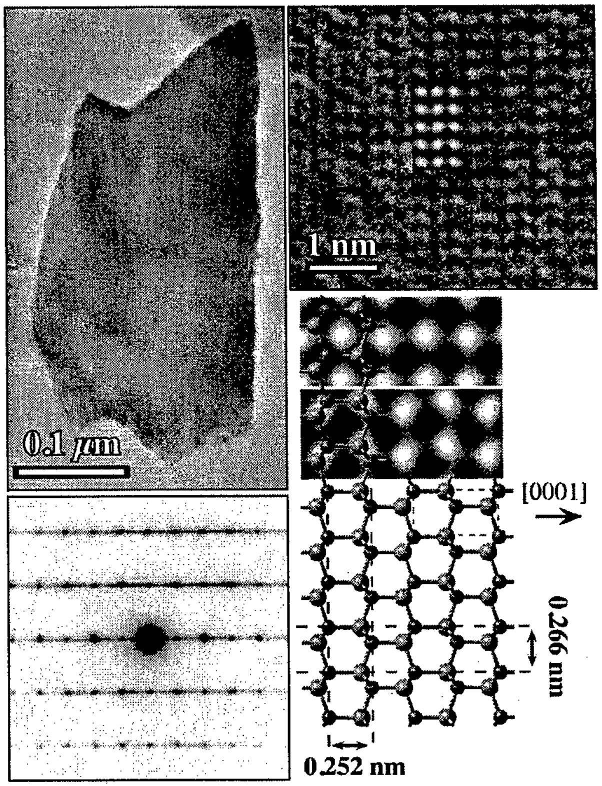

Figure 2. Bright-field image, HR-TEM lattice image, and SAED pattern for a 2H α-SiC in Murchison KJB. An atomic model for the [11-20] zone axis is shown superimposed on simulated HR-TEM images at two defocus values, the topmost simulation matches the imaging conditions of the HR-TEM image.

There are inherent difficulties in determining relative abundances of grain types using SAED and high resolution (HR)-TEM images. Difficulties arise because of the finite tilt range of the TEM goniometer and from the fact only two dimensional crystallographic information is contained in any one combination of SAED pattern and HR-TEM image. To identify polytype, a grain must be oriented to a high symmetry zone axis perpendicular to the tetrahedral stacking direction. Because of the finite tilt limits of the goniometer, a fraction of the randomly oriented grains will have no suitable high symmetry zone axes accessible. This fraction varies with SiC polytype. Nonetheless, the actual distribution can be estimated by applying appropriate corrections to TEM measured distributions (Table 1). This was accomplished by calculating, ε_i, the intrinsic fraction of randomly oriented crystals having at least one suitable zone axis (i.e., perpendicular to the tetrahedral stacking direction such as cubic <011> or hexagonal <11-20>) within the TEM goniometer tilt limits. In addition to crystal symmetry, ε_i is also dependent on twin and polytype-intergrowth microstructure. Both were taken into account in calculations used to correct the distributions.

Table 1: Murchison KJB SiC Polytype Distribution
Based on TEM analysis of 107 Grains

Grain Type	Population (%)	
3C	82.42 ± 1.95	
2H/3C	11.57 ± 2.64	16.09 ±2.64
2H	4.52 ± 0.08	
Disordered	1.50 ± 0.03	
All other polytypes	< 1%	

As demonstrated here, the KJB separate contains a large number of SiC grains containing 2H structure (16.09 ± 2.64 %) as in intergrowths and single crystals. In light of the bulk and individual isotopic data [16-19], together with the abundance of these grains, it is difficult to attribute all of them to nebular products. Therefore, 2H must be a presolar SiC grain type. The occurrence of two polytypes and their intergrowths indicates presolar SiC formed under a wider range of conditions than previously thought.

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